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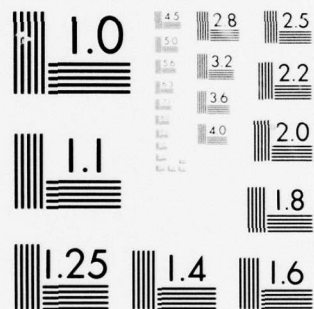
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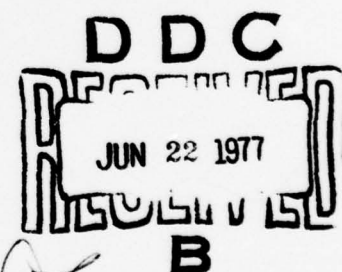
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TECHNICAL REPORT AFFDL-TR-76-129  
FINAL REPORT FOR PERIOD MAY 1975 - OCTOBER 1975

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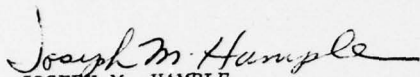


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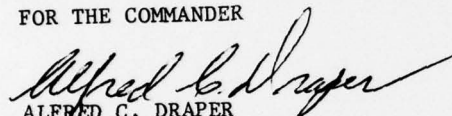
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This Technical Report has been reviewed and is approved for publication.

  
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)  Selective catalytic reduction, using ammonia as the reductant over a zeolyte catalyst, was chosen for the abatement of nitrogen oxides from the Facility (Technical Report AFFDL-TR-75-46). Equipment design specifications were prepared for a system consisting of: (a) two activated carbon adsorption beds, (b) two fluidized bed catalytic reactors, (c) bypass and emergency relief, and (d) temperature/ammonia controls. The activated carbon beds remove oil from the Facility reciprocating compressors to eliminate the potential of explosion with ammonium nitrate in the reactor. (cont'd on reverse)			

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20. ABSTRACT (cont'd).

→ The fluidized bed reactor combines nitrogen oxide containing gas with ammonia producing nitrogen and water with less than 100 ppm residual nitrogen oxides. The reactor operates at 350-400°C and uses the exothermic heat of reaction of nitrogen oxides and oxygen with ammonia to heat the 80°C inlet gas and ammonia. The bed is jacketed with commercial heat transfer oil to remove excess heat. ↗

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# FOREWORD

This report was prepared by RRC International, Inc. of Latham, New York and summarizes their work under the second phase of Contract No. F33615-74-C-3076. Mr. Joseph M. Hample of the Air Force Flight Dynamics Laboratory (AFFDL/FXN) was Project Engineer. He was assisted by Mr. Perie R. Pitts, Jr. (AFFDL/FXN).

The work summarized in this report was initiated in May 1975 and completed in October 1975 at which time this report was released.

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## SECTION I

### SUMMARY

RRC International, Inc. has investigated methods for the abatement of nitrogen oxides from the effluent of the 50 MW Hypersonic Test Leg (HTL) Facility of the U. S. Air Force Flight Dynamics Laboratory (AFFDL). The first phase of this program involved the technical and economic evaluation of potential abatement schemes. The second phase involved the formulation of design criteria for the abatement method selected in the first phase. This is the report for the second phase.\*

The abatement method recommended by RRC and accepted by AFFDL was selective catalytic reduction by ammonia with a zeolite catalyst. This method was chosen because it would produce a very clean effluent gas at relatively low cost. Because of the unique operating conditions of the HTL and high  $\text{NO}_x$  levels, it was necessary to design a complete system specifically for this application. The system, with an ultimate capacity of 10 lb/sec flow, 2 vol %  $\text{NO}_2$ , 1 vol %  $\text{NO}$ , consists of the following: (a) two activated carbon adsorption beds, (b) two fluidized bed catalytic reactors, (c) bypass and emergency relief, and (d) temperature/ammonia controls. Specifications for this equipment have been formulated and are tabulated in the appropriate report sections.

The activated carbon beds remove oil from the reciprocating compressors to eliminate the potential of explosion with ammonium nitrate in the reactor. The fluidized bed reactor combines  $\text{NO}_x$  containing gas with  $\text{NH}_3$  producing  $\text{N}_2$  and  $\text{H}_2\text{O}$  with less than 100 ppm residual  $\text{NO}_x$ .

\* Note: A report for Phase I has been issued as Technical Report AFFDL-TR-75-46.



The reactor is at 350-400°C and uses the exothermic heat of reaction of NO, NO<sub>2</sub> and O<sub>2</sub> with NH<sub>3</sub> to heat the 80°C inlet gas and inlet NH<sub>3</sub>. The bed is jacketed with commercial heat transfer oil to remove excess heat.

The fluidized bed was chosen because a packed bed doesn't provide adequate heat transfer capability. Transfer coefficients are 6-10 times greater for fluidized beds. By using a fluidized bed, other benefits are realized. Temperature uniformity is excellent, with, at most, a 10°F difference from point-to-point above the first 3" of the bed. This rapid attainment of temperature uniformity precludes the need for separate inlet gas preheaters. Besides eliminating expensive preheater hardware, the heating of inlet gas is a free and convenient sink for the exothermic heat of reaction. The heating of entering NH<sub>3</sub> is another heat sink. The combination of these sinks reduces the heat load from  $9.28 \times 10^6$  BTU/HR for 2 vol % NO<sub>2</sub>, 1 vol % NO and 50% excess NH<sub>3</sub> at 10 lb/sec flow to a maximum of  $3.74 \times 10^6$  BTU/HR; the excess is transferred through the bed walls to oil which heats from 140-240°F. The oil is cooled by water and returned to the reactor. Water is available at 85°F and exits at 125°F at a flow of 190 gpm. Oil flow is 170 gpm.

A flow of 10 lb/sec requires two fluidized beds each with a 5 lb/sec capacity. Two beds each containing 51 1/2 ft<sup>3</sup> of 20-30 mesh Zeolon 900H aggregate are required to accommodate the maximum gas velocities and to prevent entrainment of catalyst. The smaller aggregate was required instead of 1/16" extrudate because fluidization of large pellets results in unacceptable pressure drops. The particle size of the aggregate was such that a single bed for 10 lb/sec could not be designed.



The only automatic control necessary is for  $\text{NH}_3$  flow control cutoff in the event of reactor temperatures below  $300^\circ\text{C}$ . This condition is unlikely but control is necessary for safety. Below  $300^\circ\text{C}$  ammonium nitrate formation could result in an explosion in the reactors. Even if no  $\text{NO}_x$  is present, however, the  $\text{NH}_3/\text{O}_2$  reaction (which occurs after all  $\text{NO}_x$  has reacted) will generate sufficient heat to maintain the reactor temperature above  $350^\circ\text{C}$ .

At startup, the cold unfluidized reactors will be heated in 1 hr by internal electric heaters (70 kW total) to  $220^\circ\text{C}$  at which time the beds, fluidized with air, will be heated to  $400^\circ\text{C}$  in 2 - 2 1/2 min by the  $\text{NH}_3/\text{O}_2$  reaction. The automatic  $\text{NH}_3$  shut-off must be deactivated at this time. The bed is then ready to receive any level of  $\text{NO}_x$  up to 2 vol %  $\text{NO}_2$  and 1 vol %  $\text{NO}$ .



## SECTION II

### INTRODUCTION

This report summarizes the findings of the second phase of a study conducted for the United States Air Force Flight Dynamics Laboratory (AFFDL) to determine the best method of abating a nitrogen oxides effluent from a hypersonic test facility. The study was divided into two separate phases. The first phase goal was to develop recommendations for the optimum abatement method.\* The second phase has developed definitive design criteria for the abatement method recommended by RRC in the first phase.

#### A. Statement of the Problem

The 50 Megawatt (MW) Hypersonic Test Leg (HTL) Electrogasdynamics Facility produces a high enthalpy plasma by heating air to extremely high temperatures in an electric arc. Under these conditions, nitrogen and oxygen are dissociated and recombine to form significant amounts of nitric oxide (NO). The NO enters the facility vacuum system and is partially oxidized by oxygen to nitrogen dioxide (NO<sub>2</sub>). Nitrogen dioxide and nitric oxide are both corrosive, highly oxidizing, physiologically irritating and toxic.

#### B. Proposed Abatement System

RRC has recommended and AFFDL has concurred that the most cost effective and environmentally beneficial abatement method is selective catalytic reduction by ammonia with a zeolite catalyst. This method would reduce essentially all of the NO and NO<sub>2</sub> in the effluent to nitrogen and water vapor.

---

\* Note: A report for the first phase has been issued as Technical Report AFFDL-TR-75-46.



### C. Design Parameters for Proposed Abatement System

In the design of the abatement system, the following parameters have been used:

#### 1. Inlet Conditions

Mass Flow Rate	10 lbm/sec <sup>(1)</sup>
Volume Flow Rate	7,500 SCFM (0°C, 760 mm Hg)
Temperature	80°C
NO Concentration	1 vol % maximum, 0.25 vol % minimum
NO <sub>2</sub> Concentration	2 vol % maximum, 0.25 vol % minimum
Oil Concentration	1 qt/hr <sup>(2)</sup>

#### 2. Desired Outlet Conditions

NO Concentration	50 ppm
NO <sub>2</sub> Concentration	0 ppm

#### 3. Abatement System Parameters

Pressure Drop (initial)	4 psi maximum
Pressure Drop (final) <sup>(3)</sup>	5 psi maximum

- (1) Maximum design flow for the HTL. Present heater capability: 7 lbm/sec. Normal run: about 5 lbm/sec.
- (2) Oil content is independent of mass flow rate. Oil contributed by the reciprocating compressors could present a potential hazard if ammonium nitrate were formed in the reactor.
- (3) Pressure change due to pick up of oil on activated carbon.



#### D. Special Design Considerations

The HTL is a unique facility and is operated in a manner which greatly complicates the design of any abatement system. The run times, flow rates,  $\text{NO}_x$  concentrations and periods of operations are all variable over a large range and must be considered heavily in the design. The system is operated for variable time periods ranging from just a few minutes to as long as 30 minutes but seldom over 20 minutes. The vacuum system is designed to handle 10 lbm/sec but the presently installed arc heater has a capacity of only 7 lbm/sec. Normally, most runs are 5 lbm/sec or less and almost all are presently in the 3-6 lbm/sec range. At the request of AFFDL, the abatement facility has been designed for 10 lbm/sec. Consideration has been given to the normal operating conditions by designing certain components as two parallel units of 5 lbm/sec capacity each.

The  $\text{NO}_x$  concentrations in the effluent are not well characterized. Measurements by AFFDL indicate NO in the range 5,200 to 10,400 ppm (8,100 ave.),  $\text{NO}_2$  in the range 4,400 to 19,500 ppm (10,900 ave.), and total  $\text{NO}_x$  in the range 12,800 to 24,000 ppm\*. Because of these uncertainties, the abatement process was designed for the highest possible load. This was taken as 1% (10,000 ppm) NO and 2% (20,000 ppm)  $\text{NO}_2$ . Considering the maximum flow rate and concentration, the abatement facility would require a design capacity of 0.42 lbm  $\text{NO}_x$ /sec but would usually operate at about one-third of the design capacity. There are also changes in the  $\text{NO}_x$  concentration of the gas arriving at the inlet to the abatement system during a single run (i.e., no  $\text{NO}_x$  or low  $\text{NO}_x$

\* Note: In many cases, data for both NO and  $\text{NO}_2$  for the same run are lacking and therefore the total may be higher.



at start up due to the system's residence time).

These uncertainties in inlet conditions require that the oil removal system, ammonia system, reactor and reactor cooling system be designed for maximum flow rate and maximum  $\text{NO}_x$  concentration, while inlet gas heating must account for maximum flow rate but minimum  $\text{NO}_x$  concentration.



### SECTION III

#### PROCESS DESIGN

##### A. General Overview

A flow diagram of the proposed ammonia-zeolite abatement system appears as Figure 1. The  $\text{NO}_x$  contaminated effluent from the reciprocating compressors passes through mufflers where the peak-to-peak pressure pulses are assumed to be reduced to about 0.4 psi. The effluent then passes through a bed of activated carbon which removes essentially all of the oil contributed by the compressors. The oil-free effluent enters the fluidized bed catalytic reactor and is mixed with  $\text{NH}_3$ .  $\text{NO}_x$  is selectively reduced to  $\text{N}_2$  and  $\text{H}_2\text{O}$  and any excess  $\text{NH}_3$  is oxidized to  $\text{N}_2$  and  $\text{H}_2\text{O}$ .

A bypass will allow the gas to exit from the facility without passing through the abatement system. The bypass will be used during pump-down to avoid loading the oil removal system and cooling the catalyst bed. A rupture disc will be provided to automatically vent the effluent to the bypass in case of malfunction of the abatement system.

The design of the oil removal system and fluidized beds is such that two parallel units, each capable of one-half of the maximum flow is the most economical and practical alternative.



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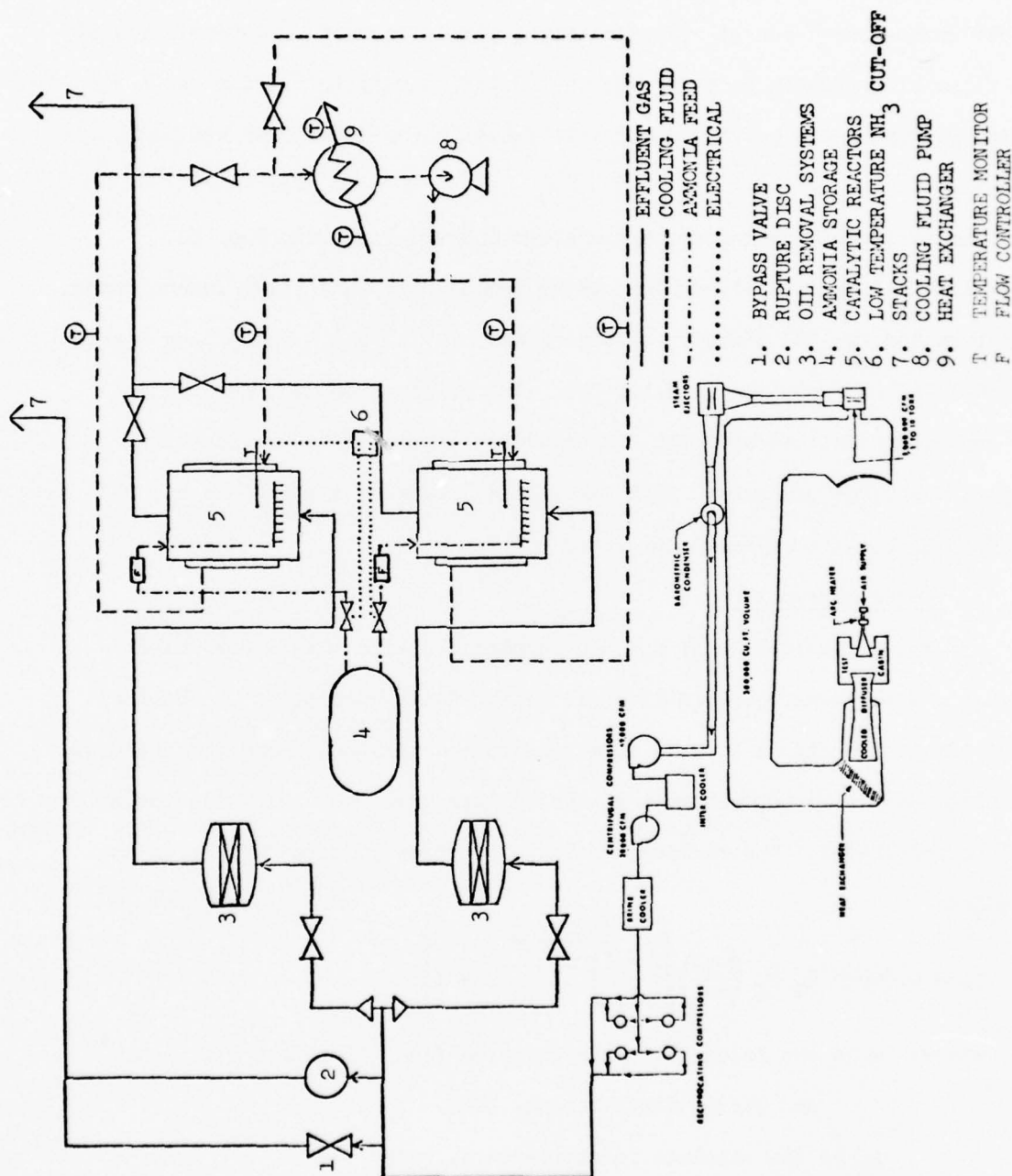


FIGURE 1. FLOW DIAGRAM OF AMMONIA ZEOLITE ABATEMENT SYSTEM



## B. Vacuum Pump Capability

The vacuum system associated with operation of the HTL is composed of two steam ejectors, centrifugal and reciprocating mechanical compressors and intercoolers. At present, the reciprocating compressors are discharging to the Smoke-X scrubber at approximately 1 psig. It must be determined what the allowable pressure drop for the  $\text{NH}_3$ /zeolite catalytic reactor and associated equipment can be in order not to exceed motor horsepower and pump capacity.

The reciprocating compressors are two Chicago Pneumatic Tool Co. (Franklin, Pa.) horizontal, duplex, water cooled, simplate valve vacuum pumps. Each pump has two 30" diameter cylinders with 12" stroke. The piston displacement of each machine is 5,430 CFM. The original design specifications for each compressor were: 2,970 CFM capacity, 1 psia inlet pressure and 18 psia discharge pressure. Each machine is driven by a 250 brake hp, 277 RPM General Electric synchronous motor.

### 1. Motor Horsepower

Mr. L. Stalzsmith of Chicago Pneumatic stated that a corrected adiabatic horsepower equation is applicable. Brake horsepower is obtained by dividing theoretical by 0.85 to get indicated horsepower and then by 0.95. Calculated brake horsepower must always be less than 250, otherwise the motors will be overloaded. Theoretical adiabatic horsepower is computed from the equation:

$$\text{hp} = 0.00436 Q_1 P_1 \left( \frac{k}{k-1} \right) \left[ \left( \frac{P_2}{P_1} \right)^{(k-1)/k} - 1 \right]$$

where:  $Q$  is the inlet flow rate computed from volumetric efficiency\*  
and piston displacement, CFM.

$P_1$  is the absolute inlet pressure, psia.

\* Chicago Pneumatic provided volumetric efficiency vs. compression ratio data.



$p_2$  is the absolute discharge pressure, psia.

and

$k$  is the ratio of specific heat of constant pressure to  
that at constant volume, 1.395 was used throughout.

At steady state operating conditions, the horsepower requirements are low. For example, for the original design conditions,  $Q_1 = 2,970$  CFM,  $p_1 = 1$  psia and  $p_2 = 18$  psia, the brake horsepower is 71.7. At startup of a vacuum system,  $p_1 = 14.7$  psia. When the calculation is performed in increments starting with  $p_1 = 14.7$ , horsepower increases from 88.2 to a maximum of 206.2 at  $p_1 = 6$  psia then decreases (see Table I).

This peaking phenomenon exists for pumpdown for any vacuum pump. Similar calculations were performed to determine the abatement equipment pressure drop,  $p_2$ , which would result in a peak horsepower equal to or less than 250. Table II gives the calculated brake horsepower for pressure drops between 4 and 10 psi (i.e.  $p_2$  between 18.7 and 24.7 psia, respectively). Above  $p_2$  of 21.7 psia, the peak horsepower requirement is greater than 250, thus, exceeding the capability of the motors. Therefore, the maximum allowable abatement system pressure drop is 7 psi.

## 2. Pump Capacity

Table III gives the available combined inlet flow capacity, for both pumps, at discharge pressures of 21.7, 19.7, and 15.7 psia for various inlet pressures. Also shown is the volumetric flow equivalent to air flows of 5 and 10 lb/sec (at 40°F), which increases as the inlet pressure is reduced. Available capacity, however, decreases with lower inlet pressure due to decreased volumetric efficiency. For each of the three discharge pressures,



TABLE I

HORSEPOWER AND CAPACITY VERSUS INTAKE  
PRESSURE AT THE ORIGINAL DESIGN OUTLET  
PRESSURE OF 18 psia FOR EACH MOTOR/PUMP

<u>INTAKE PRESSURE</u> <u><math>p_1</math>, psia</u>	<u>CAPACITY</u> <u>ACFM</u>	<u>BRAKE</u> <u>HORSEPOWER</u>
14.7	5,332	88.2
13	5,267	126.0
12	5,235	145.7
11	5,197	163.0
10	5,153	177.9
8	5,061	199.2
6	4,941	206.2
4	4,740	191.9
2	4,279	140.7
1	2,970	71.7



TABLE II

HORSEPOWER AND CAPACITY VERSUS INTAKE  
PRESSURE AT VARIOUS DISCHARGE PRESSURES  
FOR EACH MOTOR/PUMP

DISCHARGE PRESSURE <u>p<sub>2</sub>, psia</u>	INTAKE PRESSURE <u>p<sub>1</sub>, psia</u>	CAPACITY <u>ACFM</u>	BRAKE <u>HORSEPOWER</u>
24.7	14.7	5,186	230.0
	13	5,131	253.4
	12	5,093	264.2
	11	5,061	273.1
	10	5,023	279.3
22.7	8.5	4,985	259.0
	8	4,968	260.3
	7.5	4,941	260.2
22	8	4,985	252.1
	7.5	4,952	252.2
	7.0	4,941	252.5
	6.5	4,887	250.6
21.9	7.5	4,958	251.2
	7.0	4,941	251.3
	6.5	4,892	248.8
21.8	7.5	4,952	249.7
	7.0	4,941	250.1
	6.5	4,892	247.7
21.7	9.0	5,023	243.8
	8.0	4,985	248.2
	7.5	4,958	248.7
	7.0	4,930	248.4
	6.0	4,860	244.0
20.7	9.0	5,450	230.4
	8.0	5,006	235.8
	7.5	4,979	237.1
	7.0	4,952	237.4
	6.5	4,914	236.3
	6.0	4,876	234.2
19.7	14.7	5,289	128.1
	10.0	5,120	206.6
	8.0	5,023	222.6
	7.0	4,968	225.7
	6.5	4,941	225.8
	6.0	4,898	224.2
	5.0	4,811	217.5
18.7	6.5	4,958	214.2
	6.0	4,941	214.6
	5.5	4,887	212.2



TABLE III

REQUIRED AND TOTAL AVAILABLE CAPACITY  
FOR BOTH VACUUM PUMPS VERSUS INLET PRESSURE

$p_1$ (psia)	REQUIRED FLOW FOR 5 lb/sec AT 40°F (CFM)	REQUIRED FLOW FOR 10 lb/sec AT 40°F (CFM)	AVAILABLE FLOW $p_2=15.7$ psia (CFM)	AVAILABLE FLOW $p_2=19.7$ psia (CFM)	AVAILABLE FLOW $p_2=21.7$ psia (CFM)
14.7	3,775	7,550	10,806	10,578	
13	4,268	8,536	10,676		
12	4,624	9,248		10,382	
11	5,044	10,088	10,500	10,262	10,241
10	5,549	11,098	10,426	10,240	10,143
9	6,165	12,330	10,312		10,046
8	6,936	13,872		10,046	9,970
7	7,926	15,852	10,102	9,936	9,872
6	9,248	18,496	9,980	9,882	9,720
5	11,097	22,194	9,860	9,720	9,513
4	13,871	27,742	9,622		
3	18,495	36,990	9,316		



available capacity equals required capacity at inlet pressures between 5 and 6 psia for a 5 lb/sec flow and between 10 and 11 psia for 10 lb/sec.

To be conservative and avoid possible pump damage, the abatement system pressure is designed not to exceed 5 psi ( $p_2 = 19.7$  psia). The maximum power requirement is 226 hp and available capacity is sufficient for the maximum flow of 10 lb/sec. Since vacuum pump/motor capability is satisfactory, no further work is required in this area.



### C. Oil Removal System

The necessity of removing the oil added to the facility effluent by the reciprocating compressors was discussed in the previous report <sup>1</sup>. In addition to removing the oil downstream of the compressors, consideration was given to the operation of the reciprocating compressors in an oil-free mode. The compressors had been originally been built with carbon rings to be used without added lubrication, but were later converted to cast iron rings with forced oil lubrication. This change was necessitated by the presence of residual nitric acid in the compressors which caused considerable rusting between runs and premature wear of the carbon rings during subsequent operation. For these reasons, oil-free operation is not practical and removal methods had to be investigated.

Information supplied by the compressor manufacturer <sup>2</sup> and by AFFDL <sup>3</sup> indicated that the oil level was about 0.2-0.4 qt/hr and 1 qt/hr, respectively.

Equipment manufacturers thought to have commercially available equipment of the type required were contacted. The equipment available fell into the following categories:

1. Adsorption on activated carbon,
2. Coalescing,
3. Impingement,
4. Chemical combination,
5. Gas scrubbing,
6. Aftercooling.

Each manufacturer recommended one or more of the above in combination. Only adsorption on activated carbon was considered feasible because it was the



only method shown to effectively remove oil in all forms, i.e., droplets, aerosol and vapor.

The following specifications are recommended for an oil removal device.

Principle of Operation: Adsorption on activated carbon

Inlet flow rate: 7,500 SCFM

Inlet temperature: 80°C

Inlet pressure: 5 psig

Oil content of gas: 1 qt/hr

Pressure drop (clean carbon): 0.5 psi

Pressure drop (at full oil capacity): 1.5 psi

Total oil capacity: 0.5 qt

Materials of construction: carbon steel

Inlet and Outlet Connections: 24" flange

The following companies can provide the necessary equipment: (a) DeMarkus Corp., Buffalo, N.Y.; and (b) Selas Corp. of America\*, Dresher, Pa.

Information provided by DeMarkus Corp <sup>4</sup> suggests that it would be more economical to employ two units of 4,000 SCFM capacity than one larger unit. Each unit would have a capacity of 0.5 qt of oil. When the facility is operating at 5 lbm/sec or less, only one unit would be used. After each run, or after a total of about 30 minutes operating time, the carbon would be removed for disposal - regeneration is not possible. This would require a one-man hour effort and cost about \$100 for the carbon.

\* Note: Claimed to have equipment but did not respond with any details.



#### D. Preheater System

By using a fluidized bed catalytic reactor, the need for an independent preheater system is eliminated. The formation of ammonium nitrate is prevented by introducing  $\text{NH}_3$  into the hot reactor. The bed is not cooled by the  $80^\circ\text{C}$  inlet gas because the inlet gas almost immediately assumes the bed temperature.<sup>5</sup>

Preheating the bed prior to the introduction of  $\text{NO}_x$  into the reactor is required, however, in two situations; heating from a cold start and during the nominal 90 sec dead time<sup>6</sup> before  $\text{NO}_x$  generated at the arc reaches the reactor. In both cases, the  $\text{NH}_3/\text{O}_2$  reaction can provide almost all the heat required. Below  $140^\circ\text{C}$ , the degree of oxidation is insignificant but at  $215^\circ\text{C}$  the reaction goes to completion at a space velocity of  $12,500 \text{ HR}^{-1}$ .<sup>7</sup> The space velocity of HTL design at low flow for the unexpanded bed is  $4,615 \text{ HR}^{-1}$ . With increasing flow, the bed volume expands and the space velocity decreases. Thus, if the reactor is electrically preheated to about  $220^\circ\text{C}$ , the  $\text{NH}_3/\text{O}_2$  reaction will heat the bed to the desired temperature. Each reactor can be heated from  $70^\circ\text{F}$  to  $428^\circ\text{F}$  ( $220^\circ\text{C}$ ) in one hour with 70 kW of electric heaters placed in the bed. Then, with an air flow of 5 lb/sec and  $\text{NH}_3$  introduced into the fluidized reactor, the bed could be brought to  $400^\circ\text{C}$  in 2 - 2 1/2 min. This is computed from the following equation.<sup>5</sup>

$$\frac{T_{gi} - T_s}{T_{gi} - T_{so}} = \exp \left[ - \frac{\rho_g C_{pg}}{\rho_s C_{ps}} \times \frac{U_f}{(1 - \epsilon) L_{mf}} \times t \right]$$

where  $T_{gi}$  is the adiabatic gas temperature,  $947^\circ\text{C}$

$T_s$  is the desired temperature,  $400^\circ\text{C}$

$T_{so}$  is the initial bed temperature,  $220^\circ\text{C}$

$\rho_g$  is the air density,  $0.04 \text{ lb/ft}^3$



$\rho_s$  is the catalyst density, 71.4 lb/ft<sup>3</sup>  
 $C_{pg}$  is the air heat capacity, 0.25 BTU/lb-°F  
 $C_{ps}$  is the solid heat capacity, 0.25 BTU/lb-°F  
 $U_f$  is the air velocity at 5 lb/sec flow, 4.88 ft/sec at 300°C (avg.temp.)  
 $\epsilon$  is the void fraction, 0.37  
 $L_{mf}$  is the unexpanded bed height, 2 ft, and  
 $t$  is the time to get to the desired temperature, sec

At a flow of 2 1/2 lb/sec, the time is 4-5 min to reach 400°C.



#### E. Ammonia Feed System

The ammonia feed system consists of a storage tank (container in which ammonia is delivered), piping to the reactors and distribution piping within the reactor.

Piping will enter the reactor at the top of the fluidized bed and branch radially to distribute ammonia around the cross-section. The nozzles should be fitted with a strainer and turned down to prevent catalyst pellets from becoming trapped in the lines. The exhaust of the nozzles will be 6 inches from the catalyst support and will, therefore, be in the temperature uniform region of the bed.

Formation of ammonium nitrate on the nozzles is prevented because heat of reaction will transfer to the liquid ammonia, vaporize it, and heat it to the  $275^{\circ}\text{C}$  safe minimum. For a mass flow of 5 lb/sec to each reactor and a  $\text{NO}_x$  composition of 2%  $\text{NO}_2$  and 1%  $\text{NO}$ , 1.6 gpm  $\text{NH}_3$  gives 50% excess  $\text{NH}_3$ . The heat required to vaporize and heat the ammonia to  $275^{\circ}\text{C}$  is  $0.45 \times 10^6$  BTU/HR. Seventy-five 2 inch tubes of reactor length are required to provide sufficient heat transfer area. Therefore, no external heat need be provided.



#### F. Catalytic Reactor Design

The selective catalytic reduction of NO and NO<sub>2</sub> is a chemical reaction between NO, NO<sub>2</sub> and ammonia in which NO and NO<sub>2</sub> are abated by reducing them to N<sub>2</sub> on the surface of a catalyst.\* Selectivity means that NO and NO<sub>2</sub> will react with the ammonia fuel in preference to O<sub>2</sub>. Therefore, less fuel will be consumed than in non-selective methods where the fuel first reacts with oxygen.

The design of the reactor in which this process occurs requires resolution of three major problems:

- (1) provision of sufficient catalyst volume to promote sufficient conversion of NO<sub>x</sub> to N<sub>2</sub>,
- (2) geometric design to produce a pressure drop compatible with compressor limitations, and
- (3) provision for heat transfer.

Most of the design work has been performed for the average flow of 5 lb/sec. Since it is desired to have an abatement system capable of handling the maximum flow the facility pumping system is capable of delivering, data are presented for both 5 and 10 lb/sec.

##### 1. Catalyst Volume and Reaction Kinetics

The kinetic data necessary for prediction of reactor volumes are not available. Thomas and Pence have not established the mechanism by which NO and NO<sub>2</sub> are reduced by NH<sub>3</sub>. They have observed that in certain temperature ranges the product is either predominantly N<sub>2</sub> or N<sub>2</sub>O<sup>8</sup>. Their

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\* Note: The catalyst chosen for this work is Zeolon 900H (Norton Co., Akron, Ohio), a synthetic mordenite. It is crystalline aluminosilicate molecular sieve with very high chemical and thermal stability. It was chosen because of demonstrated superior ability as an effective, low cost catalyst for NO<sub>x</sub> reduction by NH<sub>3</sub> (8, 9).



work explored the reactions with levels of 5,000 ppm NO<sub>2</sub> and 500 ppm NO. Examination of their data failed to provide kinetic information sufficient for reactor design at the NO<sub>x</sub> levels, 2% NO<sub>2</sub>, 1% NO, found in the HTL effluent. However, a 27 SCFM pilot unit is being operated by Exxon Nuclear <sup>9</sup> in which NO<sub>x</sub> levels, up to 5%, are being successfully abated. Using Exxon's flow rate, reactor volume and conversion data, a reactor volume for the HTL effluent was chosen. This volume corresponds to a space velocity of 3,535 HR<sup>-1</sup>, based on flow at 70° F and 1 atm. For a maximum flow of 10 lb/sec, the catalyst volume is 140 ft<sup>3</sup>; for 5 lb/sec, the volume is 70 ft<sup>3</sup>.

The Exxon pilot unit is operated with 1/16" diameter zeolite pellets. The use of larger diameter pellets, however, reduces bed pressure drop if the same bed volume is retained. Thomas <sup>10</sup> performed experiments with a two inch deep bed and obtained the following NO<sub>x</sub> conversions with various pellet diameters:

1/8" - 40%, 1/16" - 80%, 1/32" - 99.5%, 1/64" - 99.97%.

These data indicate that pore diffusion and not chemical reaction rate controls the process. Therefore, the use of larger pellets would require larger bed volume and, while smaller pellets would reduce bed volume, overall pressure drop increases. Consequently, 1/16" pellets have been retained in the design.

## 2. Pressure Drop

Analysis of the Chicago Pneumatic vacuum pump power requirements showed that it would be desirable to maintain an overall abatement system pressure drop of less than 5 psi. Estimates of component pressure drops suggested that the pressure drop of the catalyst bed per se should be less than 1 psi.



Norton Co., the Zeolon manufacturer, stated that their procedure is to use a standard equation for pressure drop through packed\* beds<sup>11</sup>. The equation used is:

$$\Delta p = \frac{2(1-\epsilon)^{3-n}}{D_p g_c \phi_s^{3-n} \epsilon^3} \frac{G^2 L f_m}{\rho}$$

where:  $\epsilon$  is the fractional free volume

$D_p$  is the diameter of a sphere of the same volume as the particle

$g_c$  is the dimensional gravitational constant

$\phi_s$  is the shape factor

$n$  is an exponent, a function of the modified

Reynold's number,  $\frac{D_p G}{\mu}$

$G$  is the fluid superficial mass velocity per unit area

$L$  is the bed depth

$f_m$  is the friction factor, a function of the modified

Reynold's number

$\rho$  is the fluid density

$\mu$  is the fluid viscosity

Substitution of the appropriate constants for pellets of 1/16" diameter yields:

$$\Delta p = 5.9 \times 10^{-8} \frac{G^2 L f_m}{\rho}$$

where:  $\Delta p$  is in psi

$G$  is in lb/hr-ft<sup>2</sup>

$L$  is in ft

$\rho$  is in lb/ft<sup>3</sup>

For 1/8" pellet, the equation is divided by 2.

\* Note: As will be seen, the equation is useful for fluidized beds as well.



Pellet filled tubes were chosen as the basic geometry to explore the effect of the 1 psi restriction on design. Data were generated for 70 ft<sup>3</sup> of catalyst for a flow of 5 lb/sec, 1/16" diameter pellets, and a fluid temperature of 400°C, the maximum desired reaction temperature. In Table IV, the computed pressure drop is given for single and multiple tubes of varying diameter. In the case of multiple tubes, the tubes are in parallel flow arrangement so that the given pressure drop is the total bed pressure drop. It is clear from the data that the maximum allowable bed depth for a  $\Delta p$  of less than 1 psi is about 1 ft. The chart below summarizes the results for beds having the same  $\Delta p$  for flows of 5 and 10 lb/sec:

<u>Number of 1 ft Thick Beds</u>	<u>Bed Diameter for 5 lb/sec Flow</u>	<u>Bed Diameter for 10 lb/sec Flow</u>
1	8 - 9 ft	11 1/4 - 12 3/4 ft
2	6 - 7 ft	8 1/2 - 10 ft
3	5 - 6 ft	7 - 8 1/2 ft
6	3 - 4 ft	4 1/4 - 5 3/4 ft
20	< 1 1/2 ft	< 1 3/4 ft

Another configuration which satisfies the pressure drop requirements is an annular geometry. A 70 ft<sup>3</sup> catalyst bed for the 5 lb/sec average flow could be accommodated in an annulus with a 5 1/2 ft exterior diameter, a 3 1/2 ft interior diameter and a height of 5 ft. The bed depth is 1 ft and  $\Delta p = 0.71$  psi at 400°C with 1/16" diameter pellets. An annular configuration for the maximum flow of 10 lb/sec and the same  $\Delta p$  has the same diameters as above and a bed height of 10 ft. (For the 5 lb/sec case, a 70 ft<sup>3</sup> bed could have heights of 3, 10 and 20 ft with exterior diameters of 8 1/2, 3 1/4, and 2 ft, respectively.) The annular configura-



TABLE IV

CATALYST BED PRESSURE DROP FOR SINGLE  
AND MULTIPLE PACKED TUBES FOR A 70 FT<sup>3</sup>  
BED, A TOTAL FLOW OF 5 LB/SEC, 1/16 "   
DIAMETER PELLETS AT 400°C

<u>NUMBER OF TUBES</u>	<u>DIA. OF EACH TUBE (FT)</u>	<u>DEPTH OF EACH TUBE (FT)</u>	<u>OVERALL <math>\Delta p</math> (psi)</u>
1	3.0	9.9	165.70
	4.0	5.8	37.30
	5.0	3.6	12.82
	6.0	2.5	5.23
	7.0	1.8	2.72
	8.0	1.4	1.41
	9.0	1.1	0.89
	10.0	0.9	0.53
2	2.0	11.1	219.14
	3.0	5.0	26.63
	4.0	2.9	7.12
	5.0	1.8	2.58
	6.0	1.3	1.14
	7.0	0.9	0.64
	8.0	0.7	0.31
	9.0	0.6	0.19
	10.0	0.5	0.15
3	2.0	7.4	78.36
	3.0	3.3	10.52
	4.0	1.9	2.92
	5.0	1.2	1.03
	6.0	0.8	0.55
	7.0	0.6	0.25
	8.0	0.5	0.12
	9.0	0.4	0.11
	10.0	0.3	0.06



TABLE IV (Con't)

<u>NUMBER OF TUBES</u>	<u>DIA. OF EACH TUBE (FT)</u>	<u>DEPTH OF EACH TUBE (FT)</u>	<u>OVERALL p (psi)</u>
6	1.0	14.85	399.47
	1.5	6.60	315.63
	1.66	5.39	187.13
	1.75	4.85	141.90
	2.0	3.71	13.71
	2.5	2.38	4.75
	3.0	1.65	2.08
	4.0	0.97	0.61
	5.0	0.60	0.25
	6.0	0.42	0.11
	7.0	0.30	0.06
	8.0	0.23	0.04
	9.0	0.18	0.03
	10.0	0.15	0.02
20	0.50	8.91	138.10
	0.75	7.7	90.90
	0.83	6.5	62.72
	0.92	5.3	20.52
	1.00	4.5	12.82
	1.25	2.9	7.35
	1.42	2.2	4.19
	1.50	2.0	3.30

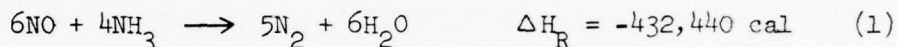


tion is preferred over tubular because it is more compact and does not require manifolded feed and exhaust connections. It will also be shown that the surface area advantage of the tubular design is insufficient for heat transfer at acceptable pressure drops.

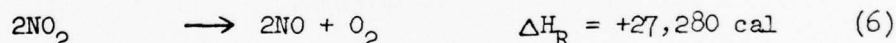
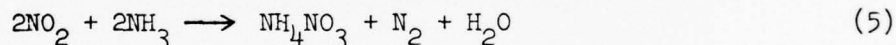
### 3. Heat Transfer

The reactions of  $\text{NO}_x$  with  $\text{NH}_3$  are exothermic. The following reactions occur at rates depending on temperature,  $\text{NH}_3/\text{NO}_x$  stoichiometry and catalyst:

#### WITH NO



#### WITH $\text{NO}_2$



#### WITH $\text{O}_2$



#### WITH $\text{N}_2\text{O}$ \*



\* Note: This reaction has been reported<sup>12</sup> on Pt catalysts at  $200^\circ\text{C}$ . It was reported to be nine times slower than reaction (2).

The heats of reaction are at  $25^\circ\text{C}$  as calculated from heats of formation at  $25^\circ\text{C}$  <sup>13</sup>. Reaction (6) occurs in the absence of  $\text{NH}_3$ .

The predominant factor determining which of the above reactions will occur is the temperature <sup>8</sup> :



1. Below 275-300°C -- Some  $\text{NH}_4\text{NO}_3$  has been detected in the effluent.
2. Between 275 and 375°C --  $\text{NO}_x$  is incompletely reduced resulting in the formation of  $\text{N}_2\text{O}$  by reactions (2) and (4). These reactions require considerably less ammonia than equations (1) and (3).
3. Below 300°C -- A considerable amount of  $\text{NH}_3$  breaks through.
4. Between 300 and 350°C -- A small amount of ammonia breaks through.
5. Between 300 and 400°C -- In this region, the catalyst will reduce  $\text{NO}_2$  to  $\text{NO}$  in the absence of ammonia. The efficiency does not become appreciable until about 350°C and reaches about 90% at 400°C.
6. Between 400 and 450°C \* -- When the catalyst is operated above 400°C, excess ammonia is oxidized to nitrogen. Ammonia, 30 to 50% in excess of that required for 99% conversion, can be employed and no significant amounts of  $\text{NO}_2$ ,  $\text{NO}$  or  $\text{N}_2\text{O}$  are found in the effluent.
7. 450°C -- At this temperature, no significant amount of  $\text{N}_2\text{O}$  is formed in gas streams even at very high water concentrations.

Based on the above data, a reactor temperature of 400°C would be required for maximum pollution reduction and for safe operation of the facility. It is particularly important that the formation of ammonium nitrates,  $\text{NH}_4\text{NO}_3$ , be avoided. The literature <sup>8, 14</sup> indicates that  $\text{NH}_4\text{NO}_3$  can be formed at temperatures as high as 330°C. This temperature is well above the normal decomposition temperature of  $\text{NH}_4\text{NO}_3$ . In actual practice, this probably represents a lack of temperature uniformity rather than the actual temperature at the site of formation. It will, therefore, be necessary to design the reactor for a high degree of temperature homogeneity.

\* Note: 450°C is the highest temperature for which data are available.



To be energy efficient, it was desired to use the heat of reaction to heat the inlet gas to reaction temperature. This would reduce the heat load in the reactor and avoid the need for additional preheat energy. From the above potential reactions, the most desirable reactions are those which produce  $N_2$  and not  $N_2O$  as a product.\* These reactions, (1), (3), and (7), are also the most energetic and require the reaction temperature to be above  $350^\circ C$ . The heat required to raise the inlet gas from  $80-400^\circ C$  at a flow of 10 lb/sec and a dew point of  $4.44^\circ C$  ( $40^\circ F$ ) is  $5.4 \times 10^6$  BTU/HR (essentially independent of  $NO_x$  content). The heat of reaction at  $400^\circ C$  calculated from heats of formation <sup>15</sup> for the same flow, 2%  $NO_2$ , 1%  $NO$  and 50% excess  $NH_3$ , is  $9.3 \times 10^6$  BTU/HR. Of this heat, 69.7% comes from the  $NO_x/NH_3$  reaction and the rest from the oxidation of excess  $NH_3$ . With a low concentration of 1/4% each of  $NO_2$  and  $NO$  the heat release from the  $NO_x/NH_3$  reaction is  $1 \times 10^6$  BTU/HR, insufficient to heat the inlet to  $400^\circ C$ . However, assuming that the same mass of  $NH_3$  is present in the high and low  $NO_x$  cases, the heat available from the excess  $NH_3$  oxidation is  $7.6 \times 10^6$  BTU/HR. For the low  $NO_x$  case, the  $NH_3$  is present to an excess of ten times the stoichiometric requirement. (The 1/2% total  $NO_x$  was deliberately chosen to be extreme.) Obviously,  $NH_3$  flow could be controlled to provide only enough heat to maintain temperature, but this means  $NO_x$  levels must be accurately known and coupled with  $NH_3$  flow control. This level of instrumental sophistication is not justified, but if desired could be attempted manually.

The transfer of the heat of reaction out of the reactor is necessary to keep the unit from overheating. Heat transfer from a packed bed is

\* Note: The formation of  $NH_4NO_3$  is considered negligible at the high operating temperatures employed and reaction (5) is not considered here.



not a great deal better than from an unpacked tube. To facilitate the required transfer, a fluidized bed reactor is necessary. The internal heat transfer coefficient is much superior to a packed bed. Additionally, a fluidized bed has a much lower pressure drop for comparable packed bed flows and is much more uniform in temperature. It is superior in transferring heat to the inlet gas; therefore, the separate preheat unit is not necessary.



### G. Fluidized Bed Catalytic Reactor

In addition to the three problems discussed for packed beds (adequate catalyst volume, pressure drop and heat transfer) the design of the fluidized bed requires consideration of minimum fluidization velocity, terminal particle velocity, and aspect ratio (bed depth to diameter).

#### 1. Catalyst Volume and Pressure Drop

As was previously stated (III.F.1.), the available data indicates that the process of abating  $\text{NO}_x$  with  $\text{NH}_3$  is controlled by pore diffusion rather than chemical reaction rate. Therefore, smaller pellets allow smaller reactor volumes. For pressure drop reasons 1/16" pellets were chosen for a fixed bed design. However, minimum fluidization requirements preclude the use of such large pellets. Zeolon 900H is available in 20 - 50 mesh aggregate and can be obtained in tighter mesh ranges. Investigation of terminal particle velocity and reactor geometry (details in III. G.3.) showed that a 20 - 30 mesh range was the best choice, but only if the maximum flow rate is 5 lb/sec. Therefore, for a maximum of 10 lb/sec two reactors are required.

Since smaller catalyst pellets are used, smaller reactor volumes are required. For a 5 lb/sec flow, 70 ft<sup>3</sup> of 1/16" pellets are needed. Assuming that for each halving of catalyst pellet diameter, half the reactor volume is required, 20 - 30 mesh pellets (equivalent particle diameter 0.034 in) required 39 1/2 ft<sup>3</sup> for 5 lb/sec. However, due to backmixing within the fluidized bed, larger reactor volumes are needed for high chemical conversions. Assuming 30% extra required volume, the total reactor volume is 51 1/2 ft<sup>3</sup>.

The equation previously used for pressure drop calculations in packed beds is applicable for fluidized beds by substituting mass flow



per unit area at the minimum fluidization velocity for G in the equation. For the 51 1/2 ft<sup>3</sup> bed of 20 - 30 mesh Zeolon and a fluidization flow of 1/2 lb/sec, the total pressure drop is 0.24 psi at 350°C.

## 2. Minimum Fluidization Velocity, Terminal Velocity and Aspect Ratio

The minimum fluidization velocity is the gas velocity, upward through a bed of solids, at which the frictional force between a particle and fluid counterbalance the weight of the particle; the vertical component of the compressive force between adjacent particles disappears, and the pressure drop through the bed equals the weight of fluid and particles in the bed. Terminal velocity is the upward gas velocity which exceeds the free-fall velocity of the fluidized particles. The particles are entrained and either lost or must be captured and recycled to the bed.

Calculations for minimum fluidization velocity were made with the following equation:

$$U_{mf} = \frac{d_p^2 (\rho_s - \rho_g)g}{1650 \mu_g} \quad \text{for } Re_p < 20$$

where  $U_{mf}$  is the minimum fluidization velocity

$\rho_s$  is the solid density, 71.4 lb/ft<sup>3</sup>

$\rho_g$  is the gas density

$\mu_g$  is the gas viscosity

$g$  is the acceleration of gravity

$Re_p$  is the particle Reynolds number =  $\frac{d_p U_{mf} \rho_g}{\mu_g}$

for  $\rho_g$  and  $\mu_g$  the properties of air at the appropriate temperature were used

$d_p$  is the average particle size calculated from the Norton's particle size distribution



Terminal velocity is determined by the following equation:

$$U_t = \frac{Re_{pt} \mu_g}{d_{pm} \rho_g}$$

and

$$C_d Re_{pt}^2 = \frac{4g d_{pm}^3 \rho_g (\rho_s - \rho_g)}{3\mu_g^2}$$

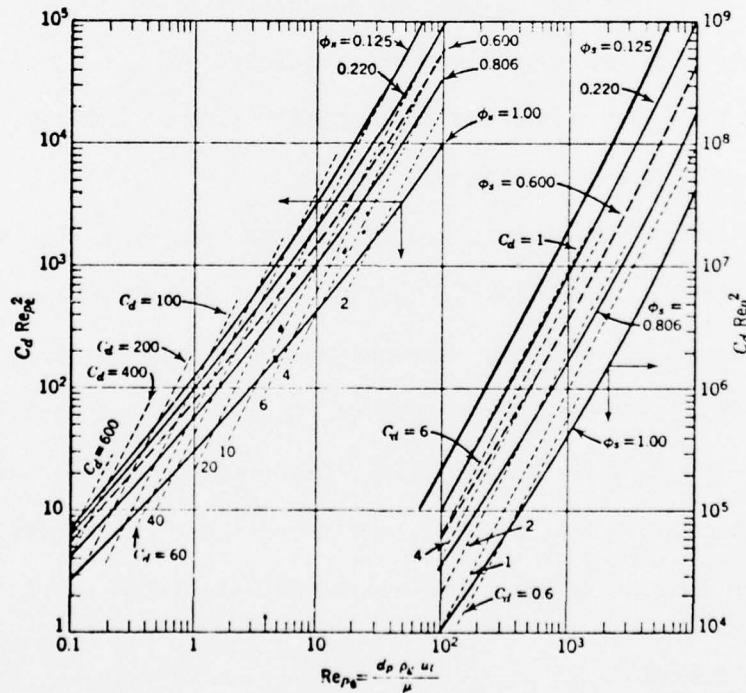
where  $d_{pm}$  is the equivalent diameter of the smallest particle

$C_d$  is an experimentally determined drag coefficient

$Re_{pt}$  is the particle Reynold's number at  $U_t$

and other symbols are as before.

Calculate  $C_d Re_{pt}^2$ , get  $Re_{pt}$  from the following chart and compute  $U_t$  from  $Re_{pt}$ .





For both terminal and minimum fluidization velocities, calculations were conducted for a bed temperature of 350°C.

Once  $U_{mf}$  and  $U_t$  are determined, the choice of minimum mass velocity which will induce fluidization is made. Whether or not this choice is acceptable depends on anticipated minimum test facility mass flows, the bed height to bed diameter ratio and if the anticipated maximum mass flow produces a gas velocity greater than  $U_t$ . The trend is that the greater the chosen mass flow for fluidization, the greater the bed diameter and the more shallow the bed, while an aspect ratio close to unity is desired. It will always produce a velocity greater than  $U_t$  if the minimum mass flow for fluidization is not above a predictable minimum based on the  $U_t$  to  $U_{mf}$  ratio.

Table V gives data for  $d_p$ ,  $d_{pm}$ ,  $U_{mf}$  and  $U_t$  for various bed particle size distributions. The last two columns give the minimum mass flow for fluidization which is required in order not to violate the  $U_t$  criterion. It is given for the two cases where the maximum mass flow to a single fluidized bed reactor is 5 and 10 lb/sec.

Table VI gives the bed dimensions and maximum gas velocity,  $U_f$ , for several chosen fluidization mass flows. In each case, the chosen mass flows are above the required minimum. The unexpanded bed height is calculated from the required catalyst volume for the particular particle size distribution. As explained in III.G.1., these reactor volumes are dependent on average catalyst pellet size, and therefore are different for each size distribution. Expanded bed height is calculated from:

$$\frac{L_f - L_{mf}}{L_{mf}} = \frac{0.0188}{\sqrt{d_p}} (U_f - U_{mf})$$



TABLE V

FLUIDIZATION DATA FOR VARIOUS CATALYST  
 PELLET SIZE DISTRIBUTIONS AT 350°C

Particle Size Distribution (U.S. Sieve No.)	$d_{pm}$ (inches)	$d_p$ (inches)	$U_{mf}$ (ft/sec)	$U_t$ (ft/sec)	Minimum Mass Flow for Fluidization for Max. Flow to a Single Reactor at -	
					<u>5 lb/sec</u> (lb/sec)	<u>10 lb/sec</u> (lb/sec)
20-50	0.0117	0.02509	0.302	2.35	0.64	1.29
20-30	0.0234	0.0340	0.552	5.87	0.47	0.94
30-50	0.0117	0.0216	0.223	2.35	0.47	0.95
30-40	0.0165	0.0242	0.281	4.16	0.34	0.68
40-50	0.0117	0.0172	0.141	2.35	0.30	0.60
20-40	0.0165	0.0289	0.401	4.16	0.48	0.96



TABLE VI

BED GEOMETRY FOR VARIOUS CATALYST PELLET SIZE  
DISTRIBUTIONS AND FLUIDIZATION MASS FLOWS AT 350°C

Particle Size Distribution (U.S. Sieve No.)	Fluidization Mass Flow (lb/sec)	Bed Dia. (ft)	Unexpanded Bed Ht. (ft)	$U_f$ (ft/sec)	Expanded Bed Ht. with Max. Mass Flow at 5 lb/sec (ft)
20-50	0.65	8.79	0.63	2.32	1.15
20-30	0.5	5.71	2.01	5.52	5.54
30-50	0.5	8.99	0.51	2.23	0.89
30-40	0.5	8.02	0.73	---	---
	0.4	7.17	0.91	3.51	---
	0.35	6.71	1.04	4.01	2.66
40-50	0.5	11.36	0.26	1.41	---
	0.3	8.76	0.43	2.35	1.10
20-40	0.5	6.70	1.25	4.01	2.98



where  $L_f$  is the expanded bed height at  $U_f$   
 $L_{mf}$  is the unexpanded bed height  
and the other symbols have been explained

Of the data presented, the only catalyst bed which has a reasonable aspect ratio is the 20-30 mesh case. It is also only acceptable if a maximum of 5 lb/sec is delivered to the reactor. No case could be found for a bed capable of receiving 10 lb/sec. This can be inferred from the high minimum mass flow required for 10 lb/sec maximum as shown in Table V. Several calculations were made, however, to verify this inference.

### 3. Heat Transfer

The previous discussion concludes that two fluidized catalytic beds are required to accommodate a maximum mass flow of 10 lb/sec. Each bed contains  $51\frac{1}{2}\text{ ft}^3$  of 20-30 mesh Zeolon 900H. The diameter of each is  $5\frac{3}{4}\text{ ft}$  and the bed depth is 2 ft at or below a mass flow of  $1\frac{1}{2}\text{ lb/sec}$ . At the maximum flow of 5 lb/sec, the bed expands to  $5\frac{1}{2}\text{ ft}$  deep.

The surface area available for heat transfer varies as the bed expands with increasing mass flow. For example, at the design temperature,  $350^\circ\text{C}$ , the bed depth is  $5\frac{3}{4}\text{ ft}$  at 5 lb/sec, but at  $2\frac{1}{2}\text{ lb/sec}$ , the bed only expands to  $3\frac{1}{2}\text{ ft}$ . To determine if these surface areas are adequate to transfer the heat of reaction, the inside wall heat transfer coefficient is calculated.

Six correlations <sup>16</sup> were used to compute the heat transfer coefficient. Values ranged from 10.5 to 457 BTU/HR-ft<sup>2</sup>-°F. By comparison for the packed unfluidized annular bed, three correlations <sup>17</sup> for wall heat transfer coefficient for fixed beds gave values of less than 1 to 70 BTU/HR-ft<sup>2</sup>-°F. A conservative value of 50 BTU/HR-ft<sup>2</sup>-°F was chosen from the four most



consistent fluidized bed correlations for the maximum flow of 5 lb/sec. The range for a low flow of 2 1/2 lb/sec is 8 to 142 BTU/HR-ft<sup>2</sup>-°F using five of six fluid bed correlations (eliminating the equation which gave the high extreme value). The flow, 2 1/2 lb/sec, is not arbitrary but is the minimum facility test flow indicated by past AFFDL data. It is necessary to verify that heat transfer can be successfully achieved over the whole range of test possibilities. Again, 50 BTU/HR-ft<sup>2</sup>-°F was chosen as a conservative value.

Since the reactor can operate between 350 and 400°C, the effect higher temperature on heat transfer was checked. With the five correlations used for the 2 1/2 lb/sec evaluation, the coefficient range is 9-85 BTU/HR-ft<sup>2</sup>-°F at 5 lb/sec and again 50 was used.

Assuming that heat transfer oil would be used in a shell external to the fluidized bed, an outside wall heat transfer coefficient of 200 BTU/HR-ft<sup>2</sup>-°F was assumed. Thus, an overall coefficient, U, of 40 is computed from the equation:

$$U = \left[ \frac{1}{h_w} + \frac{1}{h_o} \right]^{-1}$$

where  $h_w$  and  $h_o$  are the inside and outside wall coefficients, respectively, and wall resistance is neglected. Also assuming that oil will enter the heat exchange area at 140°F and exit at 240°F, Table VII gives the bed heat transfer capabilities calculated from:

$$Q = U A \Delta T_{ln-m}$$

where Q is the possible heat transferred BTU/HR  
 U is the overall heat transfer coefficient  
 A is the heat exchange area, ft<sup>2</sup>, and  
 $\Delta T_{ln-m}$  is the log-mean temperature difference



TABLE VII

## HEAT TRANSFER CAPABILITY AND REQUIREMENTS

Mass Flow lb/sec	Bed Temp. °C	Sensible Heat Trans. to Heat from 80°C to Bed Temp. (10 <sup>6</sup> BTU/HR)	Heat of Reaction for 2% NO <sub>2</sub> and 1% NO and 50% Excess NH <sub>3</sub> (10 <sup>6</sup> BTU/HR)	Heat Trans. to Preheat NH <sub>3</sub> (10 <sup>6</sup> BTU/HR)	Heat of Reaction Minus Sens. Heat Minus NH <sub>3</sub> Preheat (10 <sup>6</sup> BTU/HR)	Bed Heat Trans. Capability (Oil in at 140°F out at 240°F) (10 <sup>6</sup> BTU/HR)
5	350	2.32	4.64	0.45	1.87	1.87
5	400	2.72	4.64	0.45	1.47	2.39
2 1/2	350	1.16	2.32	0.23	0.93	1.21



Table VII also gives the heat transfer requirements for the heats of reaction. Since about half of the heat generated by reaction will be transferred to the cold ( $80^{\circ}\text{C}$ ) inlet gas, the net heat to be removed is much lower than if the gas were independently preheated. The ability of the fluidized bed to rapidly transfer heat to inlet gas is responsible for this benefit.

The last two columns of the table are compared to see if transfer capability matches requirement. In all three cases, capability equals or exceeds requirements.

#### 4. Reactor Specifications

There will be two fluidized bed reactors, each capable of accommodating a maximum flow of 5 lb/sec. A sketch appears as Figure 2.

Catalyst Volume	51 1/2 ft <sup>3</sup>
Catalyst Type	20-30 mesh Zeolon 900H*
Reactor Diameter	5 3/4 ft
Unexpanded Bed Depth	2 ft
Maximum Expanded Bed Depth	5 1/2 ft
Inside Wall Heat Transfer Coefficient	50 BTU/HR-ft <sup>2</sup> °F
Reactor Temperature	350-400°C
Overall Reactor Ht. with Freeboard	8 ft

Catalyst loading and inspection sampling is through a porthole at the top of each reactor. Catalyst removal requires removing the top of the reactor and hoisting the catalyst basket and  $\text{NH}_3$  distribution tubes out of the reactor. This is only required if inspection sampling indicates catalyst

\* Note: The distribution is such that 8% of the pellets are either larger than 20 mesh or smaller than 30 mesh.



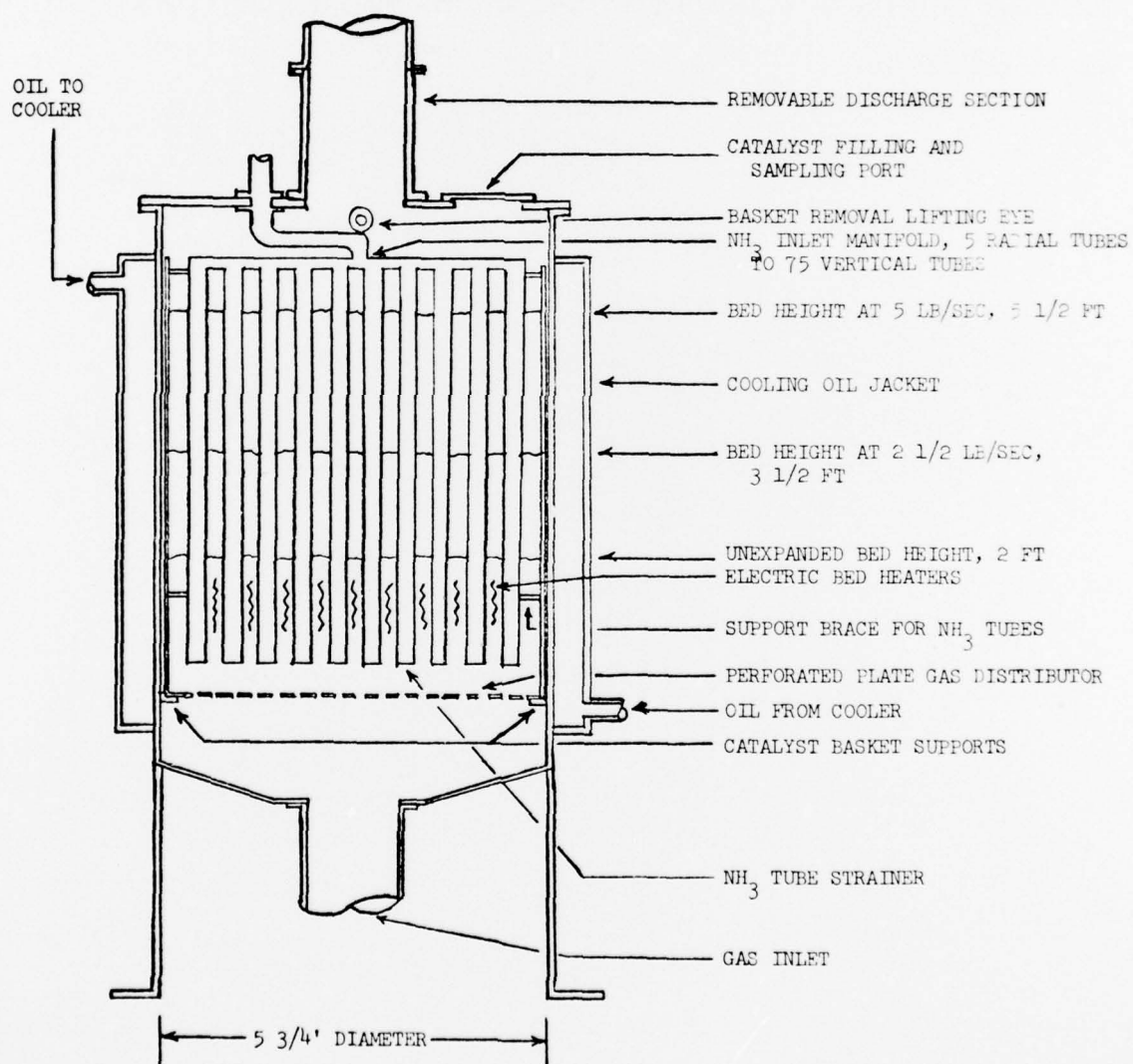


FIGURE 2. FLUIDIZED BED DETAIL



attrition or abatement performance becomes insufficient. The reactor, catalyst basket,  $\text{NH}_3$  inlet tubes, oil jacket should be constructed of 304 S.S. The  $\text{NH}_3$  tubes should preferably be seamless and the discharges be fitted with strainers. The 18" O.D. discharge pipe and exhaust valve should also be 304 S.S. There should be a high temperature, low conductivity gasket between the reactor and the inlet and outlet pipes. The inlet pipe may be plain carbon steel and the existing 24" stack may be used as is.



#### H. Heat Transfer Oil Specifications

Type	Commercial Quality
Maximum Heat Transferred for 10 lb/sec Flow	$3.74 \times 10^6$ BTU/HR
Inlet Temperature	140°F
Outlet Temperature	240°F
Circulation Rate	170 gpm
Heat Exchanger Area Required to Cool Oil	210 ft <sup>2</sup> *
Water to Heat Exchanger	
Inlet Temperature	85°F
Outlet Temperature	125°F
Circulation Rate	190 gpm

\* Note: Assumes an overall coefficient of 300 BTU/HR-ft<sup>2</sup>-°F



## I. Control and Monitoring

Provision should be made to monitor reactor bed temperature, oil and water inlet and outlet temperatures,  $\text{NH}_3$  flow rate,  $\text{NO}_x$  inlet and outlet concentrations. If the bed temperature drops below  $300^\circ\text{C}$  during a run, the  $\text{NH}_3$  flow should be automatically shut-off to prevent combining  $\text{NH}_3$  and  $\text{NO}_2$  to form explosive ammonium nitrate. Should the bed cool to below  $300^\circ\text{C}$ , the electric heaters can be brought on-line.

Other controls for preventing the bed temperature from exceeding  $400^\circ\text{C}$  can be the manual control of oil and cooling water flows as well as the limiting of excess  $\text{NH}_3$ .



#### J. Bypass and Safety Relief

A bypass will be incorporated into the abatement system to (a) permit pumpdown without passing the gas through any component of the abatement system, and (b) allow for emergency venting of the effluent in case of high back pressure in the abatement system. During pumpdown, a valve to the bypass will be open allowing the effluent to leave the facility without entering the abatement system or the presently used stack. This will (a) greatly reduce the load on the oil removal system, (b) allow for heating of the catalyst beds, (c) reduce the back pressure on the vacuum system. A separate line containing a rupture disc will circumvent the main valve to protect against over pressure.

The following specifications are recommended for the rupture disc:

Burst Pressure Rating (Maximum)	16 psig
(Minimum)	14 psig
Relief Capacity	7,500 SCFM Air
Temperature Capability	20-150°C
Materials of Construction	Impervious Graphite
Vacuum Support	(Consult Manufacturer)
Carrier and Gaskets	As necessary to fit 24" 150 lb ASA flanges.

Specify that the rupture disc meet requirements of 1968 ASME Unfired Pressure Vessel Code, Section VIII, Par. UG-127 (b) and (c).

Impervious graphite was chosen over metals because of its greater resistance to corrosive chemicals and greater temperature and pressure stability.



## SECTION IV

### REFERENCES

1. Phase I Report, AFFDL-TR-75-46, p. IV-39.
2. Private Communication, Larry Stalzsmith, Chicago Pneumatic Tool Co., August, 1975.
3. Private Communication, Perie Pitts, Jr., AFFDL, August 19, 1975.
4. Private Communications, Robert Lask, Demarkus Corporation, August 1975.
5. Kunii, D. and O. Levenspiel, Fluidization Engineering, John Wiley and Sons, Inc., New York, N.Y., 1969, pp. 420-1.
6. Private Communication, Joseph Hample, AFFDL, May, 1974.
7. Skvortsov, G. A., et al, Soviet Chem. Ind. No. 6, 45-8 (1970).
8. Pence, D. T. and T. R. Thomas, "NO<sub>x</sub> Abatement at Nuclear Processing Plants", Proceedings of the Second AEC Pollution Control Conference, Albuquerque, N.M., April 16-19, 1974, WASH-1332(74), Vol. I.
9. Mays, E. B. and M. R. Schwab, "Elimination of NO<sub>x</sub> Fumes", Chem. Eng. Feb. 17, 1975.
10. Private Communication, Thomas Thomas, Allied Chemical Corporation, August, 1975.
11. Boucher, D. F. and G. E. Aloes, Perry's Chemical Engineers' Handbook, 4th ed., R. H. Perry, et al, Ed. McGraw Hill Book Company, New York, N.Y., 1963, pp. 5-49 through 5-52.
12. Otto, K, M. Shelef, and J. T. Kummer, J. Phys. Chem., 74, 2690-2698 (1970).
13. Liley, P. E., et al, Perry's Chemical Engineers' Handbook, 4th ed., R. H. Perry, et al, Ed., McGraw Hill Book Company, New York, N.Y. 1963, pp. 3-139.
14. Ganz, S. N. and A. M. Vashkevich, Khim. Technol. No. 13, 72-6 (1968).
15. Wicks, C. E. and F. E. Block, Bureau of Mines Bulletin 605, Washington, D.C., 1963.
16. Kunii, D. and O. Levenspiel, Fluidization Engineering, John Wiley and Sons, Inc., New York, N.Y., 1969, pp. 268-272.
17. Perry, R. H., et al, Perry's Chemical Engineers' Handbook, 4th ed., R. H. Perry, et al, Ed., McGraw Hill Book Company, New York, N.Y., 1963, pp. 4-24.